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The Variability of Delivered Dose of Aerosols with the Same Respirable Concentration but Different Size Distributions

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The influences of aerosol size distribution and breath tidal volume on respirable dose estimates were examined for mouth breathing using the ACGIH/ISO/CEN criterion for respirable-equivalent aerosols. Actual tissue doses predicted from a set of pulmonary empirical deposition equations, the Heyder–Rudolf equations, were compared with deposition assumed to occur under the penetration-based respirable dust sampling criterion. Deposition estimate errors ranged from ~1/10- to 10-fold, with aerosol mass median aerodynamic equivalent diameter and geometric standard deviation as well as tidal volume each showing a substantial influence under appropriate conditions. These findings demonstrate that reliance on respirable aerosol sampling data obtained with devices performing on a penetration-based sampling criterion may lead to erroneous dose–response relationships in exposure standard development as well as exposure misclassification errors during epidemiological studies. A more reliable dose estimate would be obtained using devices with collection efficiency performance closely matching the alveolar deposition prediction curves of Heyder and Rudolf. We believe that if it is not currently required, the development of a deposition-based aerosol sampling methodology will soon be required for the determination and quantification of inhaled aerosol-induced adverse health effects.

Keywords: error; misclassification; respirable; size-selective sampling

INTRODUCTION

Ideally, air sampling devices intended to characterize occupational exposures to potentially hazardous aerosols should yield measurement results representative of a risk-related quantity that can be directly and unambiguously interpreted. The undeniable importance of considering particle size distribution as a modifier for inhaled particle-induced health risks has been a part of exposure estimation and risk characterization processes since the development of early pre-separators for sampling the 'respirable' fraction of aerosols (see for example Wright, 1954; Hyatt, 1960). Hitherto commonly employed pre-separator-based measurement methods are, one way or another, related to the probability of penetration of particles to the respiratory tract. While penetration-based measurements were a significant improvement over measurements that did not include some degree of scaling based on particle size, the difference between penetration to a specific site and deposition at that site is bound to create discrepancies in risk estimation. It has been reported that this limitation has always been clearly understood by developers of size-selective sampling criteria derived from pre-separator performance and known to closely represent, but not exactly predict, respiratory tract deposition (Walton and Vincent, 1998). In fact, whether the attribution of risks should be based on a penetration-based or a deposition-based index has recently been debated (Soderholm and McCawley, 1990; Hewett, 1991). McCawley (1993) estimated a range of differences of as much as 400% in deposition- and penetration-based dose estimates. Fisher and McCawley (1997) showed a broad range of particle size distributions that contained both sub-micrometre and larger particles from published ambient air particle size distributions, suggesting that this broad range could

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contribute to significant differences between penetration- and deposition-based dose estimates. These concerns notwithstanding, a systematic investigation of the magnitude of potential error in penetration-based dose estimation has not been reported.

Corresponding to the current aerosol sampling technology, penetration-based samplers widely used in occupational hygiene include the 10 mm cyclone and IOM personal samplers, which measure the respirable and inhalable aerosol fractions, respectively. The sampling performance of these devices conforms closely to current criteria for size-selective sampling (Soderholm, 1989; ACGIH, 2001). Studying one type of instrument would be consequential in understanding the error behaviour in all penetration-based instruments. We chose respirable aerosol sampling purely on the basis of mathematical convenience of the calculations. The method used in this paper is directly transposable to any aerosol penetration-based method of estimating aerosol dose. The ACGIH/ISO/CEN criterion for respirable aerosol samplers may be expressed as (Soderholm, 1989):

$$SR(d) = SI(d)[1 - F(x)] = SI(d)P(d) \quad (1)$$

where $SR(d)$ is the fraction of total airborne particles of size d that is expected to penetrate to the pulmonary spaces, $SI(d)$ is the fraction of total airborne particles of size d that will be inhaled and $F(x)$ is the cumulative probability function of the standardized random variable x:

$$F(x) = M\int_{-\infty}^{x} \frac{dx}{\sqrt{2\pi}} e^{-x^2/2} \quad (2)$$

Here $x = \ln(d/\mu_g)/\ln(\sigma_g)$, where d is again the particle size, μ_g is the geometric mass median aerodynamic particle diameter (MMAD) of the log-normally distributed aerosol and σ_g is the aerosol's geometric standard deviation (GSD). For respirable aerosol samplers the reference values of μ_g and σ_g are 4.25 μm and 1.5, respectively. Since $F(x)$ represents collection of particles by the sampler, $P(x) = [1 - F(x)]$ represents penetration of particles through the sampler.

$F(x)$ may also be expressed using the error function (*erf*) as (Soderholm, 1989):

$$F(x) = \frac{1}{2}\left[1 + erf\left(\frac{x}{\sqrt{2}}\right)\right] = \frac{1}{2}\left[1 + erf\left(\frac{\ln(d/4.25)}{\sqrt{2}\ln(1.5)}\right)\right] \quad (3)$$

where d is in μm. The error function may be approximated as (Hastings, 1955):

$$erf(y) = 1 - (1 + a_1 y + a_2 y^2 + \ldots + a_6 y^6)^{-16} + \varepsilon(y) \quad (4)$$

for $0 \le y \le \infty$ or, equivalently, $d \ge 4.25$ μm. The coefficient values are $a_1 = 0.070523078$, $a_2 = 0.0422820123$, $a_3 = 0.0092705272$, $a_4 = 0.001520143$, $a_5 = 0.0002765672$ and $a_6 = 0.0000430638$ (Hastings, 1955). For $-\infty < y \le 0$, i.e. $d \le 4.25$ μm, with the property of the error function:

$$erf(y) = 1 - erf(-y) \quad (5)$$

When $d \le 4.25$ μm, $y = 1.7439399 \ln(d/4.25)$ and $P(x) = [1 - F(x)] = 1$ within 6×10^{-3}%. Similarly, when $d \ge 25.1$ μm, $P(x) = 0$ within the same error. In solving the penetration equations numerically one may shorten the calculations if an error of up to 0.1% is accepted. This implies that $0.001 < [1 - F(x)] < 0.999$ or $0.0005 < erf(y) < 0.9995$, for which $y = \pm 3.1$. In terms of particle sizes, $x \le 0.72$ μm for $y \le -3.1$ and $x \ge 25.1$ μm for $y \ge 3.1$ would correspond to these bounds. Seven or more decimal place expression of the rational approximation equation coefficients for equation (4) is necessitated by the non-linear nature of the approximation and keeping the error bounds of the numerical integration processes within the desired limits.

For a given log-normally distributed aerosol, the mass fraction $P(x)$ penetrating through an ideal respirable aerosol sampler, i.e. one performing according to equation (1), the Soderholm criterion, may be readily calculated. In exposure assessment or stratification using sampling data from such a sampler, the implicit assumption is that this mass fraction represents the dose to pulmonary tissues. Consequently, similar 8 h time-weighted average respirable mass sampling results from different exposures to the same material, such as silica-containing mineral dust, would be considered to represent the same potential dose. However, it has been noted that this is an erroneous estimate because not all of the material penetrating to the pulmonary spaces will deposit there (Hewett, 1991; McCawley, 1993). The question arises as to the magnitude of these errors for different aerosol sizes and inhalation rate variations as the controlling parameters. In this work we have examined these errors for mouth breathing using empirical deposition equations as applied to aerosols that would be expected to yield the same respirable mass concentration when sampled with an 'ACGIH/ISO/CEN ideal sampler'.

Respirable equivalence

If aerosols *A* and *B* indicate the same respirable mass concentrations when sampled with an ideal respirable aerosol sampler, then they may be said to be respirable-equivalent aerosols according to the

ACGIH/ISO/CEN Soderholm penetration criterion. The respirable mass collected for aerosol *A* is:

$$A = C_1 \int_0^\infty \lambda_1 P(x)\,\mathrm{d}d \qquad (6)$$

and the respirable mass collected for aerosol *B* is:

$$B = C_2 \int_0^\infty \lambda_2 P(x)\,\mathrm{d}d \qquad (7)$$

where λ represents a log-normal mass distribution of particle sizes d with MMAD μ_g and GSD σ_g, C_1 and C_2 are the total mass concentrations of the two aerosols and $P(x) = [1 - F(x)]$ is as previously defined. If the aerosols are respirable-equivalent, then by definition:

$$C_1 \int_0^\infty \lambda_1 P(x)\,\mathrm{d}d = C_2 \int_0^\infty \lambda_2 P(x)\,\mathrm{d}d \qquad (8)$$

Substituting from above,

$$C_2 = \frac{\int_0^\infty \lambda_1 P(x)\,\mathrm{d}d}{\int_0^\infty \lambda_2 P(x)\,\mathrm{d}d} = \frac{\int_0^{0.72\mu\mathrm{m}} \lambda_1 P(x)\,\mathrm{d}d}{\int_0^{0.72\mu\mathrm{m}} \lambda_2 P(x)\,\mathrm{d}d} \frac{\int_{0.72\mu\mathrm{m}}^{25.1\mu\mathrm{m}} \lambda_1 P(x)\,\mathrm{d}d}{\int_{0.72\mu\mathrm{m}}^{25.1\mu\mathrm{m}} \lambda_2 P(x)\,\mathrm{d}d} C_1 \qquad (9)$$

However,

$$\int_0^{0.72\mu\mathrm{m}} \lambda_1 P(x)\,\mathrm{d}d = \frac{1}{2}\left[1 + erf\left(\frac{\ln(0.72\mu\mathrm{m}/\mu_{g_1}}{\sqrt{2}\ln\sigma_{g_1}}\right)\right] \qquad (10)$$

therefore,

$$C_2 = \frac{\frac{1}{2}\left[1 + erf\left(\frac{\ln(0.72\mu\mathrm{m}/\mu_{g_1}}{\sqrt{2}\ln\sigma_{g_1}}\right)\right] + \int_{0.72\mu\mathrm{m}}^{25.1\mu\mathrm{m}} \lambda_1 P(x)\,\mathrm{d}d}{\frac{1}{2}\left[1 + erf\left(\frac{\ln(0.72\mu\mathrm{m}/\mu_{g_2}}{\sqrt{2}\ln\sigma_{g_2}}\right)\right] + \int_{0.72\mu\mathrm{m}}^{25.1\mu\mathrm{m}} \lambda_2 P(x)\,\mathrm{d}d} C_1 \qquad (11)$$

Thus, a total mass concentration C_2 of aerosol λ_2 (μ_{g2}, σ_{g2}) is required to yield the same respirable mass sampling result as a concentration C_1 of aerosol λ_1 (μ_{g1}, σ_{g1}). Given any two such respirable-equivalent aerosols, it is possible to compare empirically derived estimates of actual tissue dose from each and to determine the influence of MMAD and GSD on the amount of error observed. Heyder and Rudolf and colleagues studied particle deposition in human subjects and developed a series of empirical equations describing deposition efficiencies in various respiratory tract regions (Heyder *et al.*, 1985, 1986; Rudolf *et al.*, 1986, 1988). The equations may be applied to both mouth and nose breathing over a range of breathing frequencies and tidal volumes. Deposition efficiency in the alveolar compartment for particles ≥ 0.05 μm is:

$$DE_A = (1 - d_N)(1 - d_L)(1 - d_B)\, V_A/V\, d_A \qquad (12)$$

where d_N is the regional deposition efficiency in the nose, d_L is the regional deposition efficiency in the larynx, d_B is the regional deposition efficiency in the tracheobronchial region, V_A is the aerosol volume coming to rest in the alveolar airways at end-inspiration, V is the tidal volume and d_A is the regional deposition efficiency for the alveolar region.

The reader is referred to the original articles for a description of the mathematical form of each of the terms, but we note here that the relevant variables include the particle aerodynamic diameter, mean volumetric flow rate, functional residual capacity, extra-thoracic dead space volume at end-inspiration and aerosol diffusion coefficient. We refer to equation (12) as the Heyder–Rudolf relationship. For the case of mouth breathing $d_N = 0$. Alveolar deposition predicted from equation (12) is compared with the ACGIH/ISO/CEN Soderholm sampling efficiency criterion in Fig. 1. It is apparent from Fig. 1 that the distribution of particle mass within the particle size range spanned by equation (1), the Soderholm criterion, should influence the indicated respirable mass concentration.

METHODS

A computer program using a structured Basic language (Microsoft QuickBasic) was written to: (i) calculate the value of C_2 given the MMAD and GSD values for two aerosols and an assumed value of $C_1 = 1$ arbitrary concentration units; (ii) calculate the masses D_1 and D_2 of the two aerosols predicted by the empirical equations for deposition in the alveolar spaces following oral breathing; (iii) compare the two mass fractions via the ratio D_1/D_2. The program employed the error function calculation scheme described above as well as a Romberg–Richardson integration algorithm (Hildebrand, 1974). Calculations were performed for a breathing frequency of 15 min^{-1}, tidal volumes of 750, 1500 and 2100 cm^3, μ_{g1} and μ_{g2} = 0.1, 0.2, 0.5, 1, 1.5, 2 and 5 μm, σ_{g1} = 1.5 and 3 and σ_{g2} = 1.5, 1.75, 2, 2.5 and 3. Errors were calculated as

$$\Delta\,(\%) = 100 \times (D_1/D_2 - 1) \qquad (13)$$

RESULTS AND DISCUSSION

Figures 2–4 illustrate the influences of differences in MMAD, GSD and tidal volume on respirable dose for two aerosols that would be considered respir-

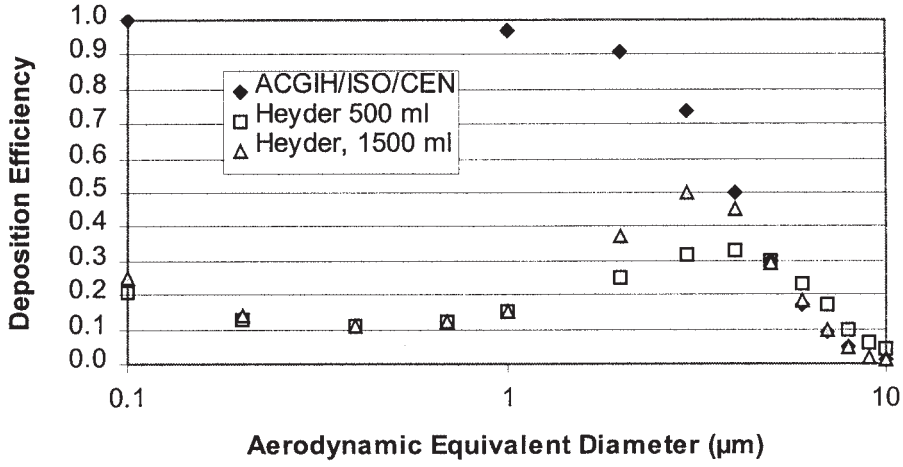

Fig. 1. ACGIH/ISO/CEN Soderholm respirable sampler efficiency criterion versus empirically derived alveolar regional deposition efficiencies. The Heyder–Rudolf empirical curves are for conditions of 500 and 1500 ml tidal volume and 15 min^{-1} breathing rate.

able-equivalent according to the ACGIH/ISO/CEN Soderholm sampling criterion, i.e. two aerosols that would provide the same collected mass during sampling. The ranges of error values for each condition are presented in Table 1.

The error range was maximum for the condition of smallest tidal volume (750 ml) and smallest GSD (1.5) for both aerosols. This range represented an ~10-fold difference in the mass depositions of the two aerosols, which under the ACGIH/ISO/CEN Soderholm sampling criterion are tacitly assumed to represent equal risk. The range decreased as tidal volume increased for fixed GSD values and decreased as the GSDs increased for a fixed tidal volume. The error range was minimum for the condition of largest tidal volume (2100 ml) and largest GSD (3.0) for both aerosols.

Error values were maximum (positively or negatively) when the mean size of one of the aerosols was 0.5 μm and that of the other aerosol was greater than ~5 μm. For the former aerosol the differences between deposition predicted from the ACGIH/ISO/CEN criterion and that predicted from the empirical equations are greatest about the empirical equation's minimum at ~0.5 μm and the small GSD maximizes the cumulative error associated with that aerosol. For the latter aerosol the size distribution falls primarily within the region where the ACGIH/ISO/CEN Soderholm and empirical curves merge, so that there is little difference between deposition predicted by the two functions. Thus, the disparity between the actual dose, as predicted by the Heyder *et al*. empirical equations, and the assumed dose, as predicted by equation (1), the Soderholm sampling criterion, is maximized. MMAD values different from 0.5 μm reduce the error for the former aerosol, while MMAD values smaller than ~5 μm increase the error for the latter aerosol, thereby reducing the net difference as calculated by equation (13). Similarly, agreement between the ACGIH/ISO/CEN Soderholm criterion and the empirical equations decreases with decreasing tidal volume for a given breathing frequency, as shown in Fig. 1, so that the net error calculated by equation (13) decreases with decreasing tidal volume, as reflected in Figs 2–4.

The effect of tidal volume is inversely proportional to its magnitude. The simplest way to observe this effect is to note the decline in maxima for any given size distribution from the high at 750 ml tidal volume to a low at the 2100 ml tidal volume (Table 1). The results show that for a given particle size distribution the magnitudes of errors diminish with increased tidal volume. It may also be observed that the relative change in error for a given particle size distribution as a function of tidal volume is not constant. This observation suggests a strong interaction between particle size distribution and tidal volume in affecting the magnitude of error. This is expected from the semi-empirical equations used in the calculations. As shown in the equations, all deposition rates in different respiratory tract compartments are strongly dependent on breathing volumetric flow. Although the particle–volumetric flow interactions are important in calculation of the magnitude of the errors, the general trend exhibited by these maxima is more important in interpretation of the results. The trends shown suggest that the errors tend to magnify during sedate periods. Since sedate periods are expected to dominate work, with only short bursts of intense activity, it is likely that the true errors would be closer to the maxima than the lower one shown under heavy exercise. In addition, during sedate periods nose breathing dominates. In nose breathing deposition of the sub-micrometre fraction is relatively stable in comparison to the larger fraction.

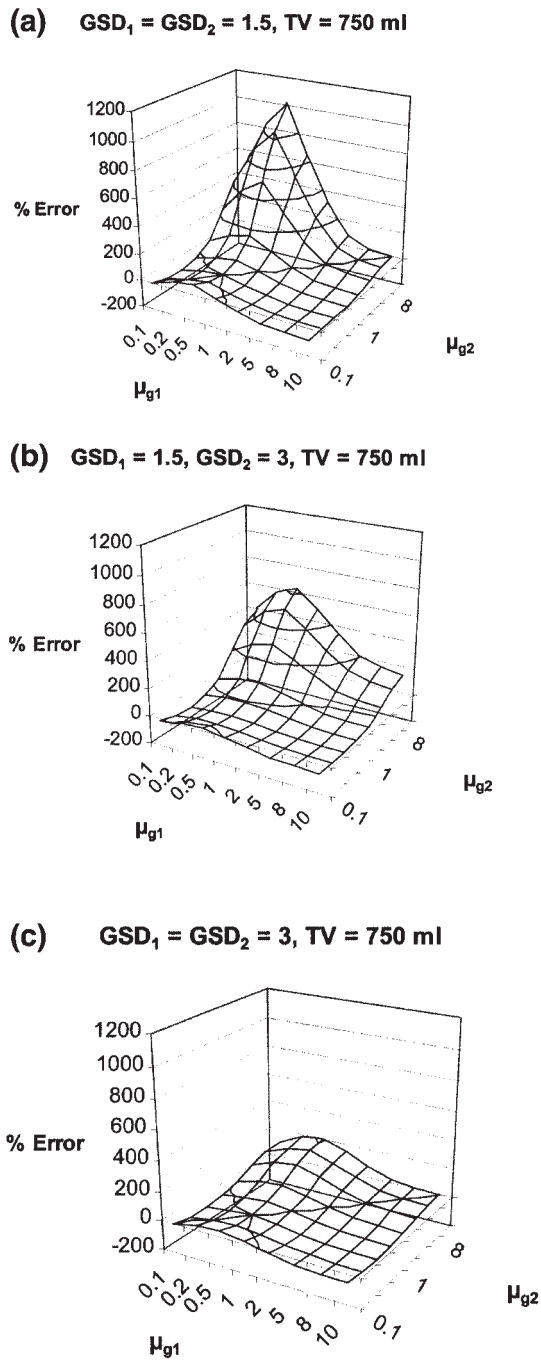

Fig. 2. Variation in percent error with μ_{g1} and μ_{g2} for breath frequency 15 min^{-1}, tidal volume 750 cm^3 and (a) $\sigma_{g1} = \sigma_{g2} = 1.5$, (b) $\sigma_{g1} = 1.5$ and $\sigma_{g2} = 3$ or (c) $\sigma_{g1} = \sigma_{g2} = 3$.

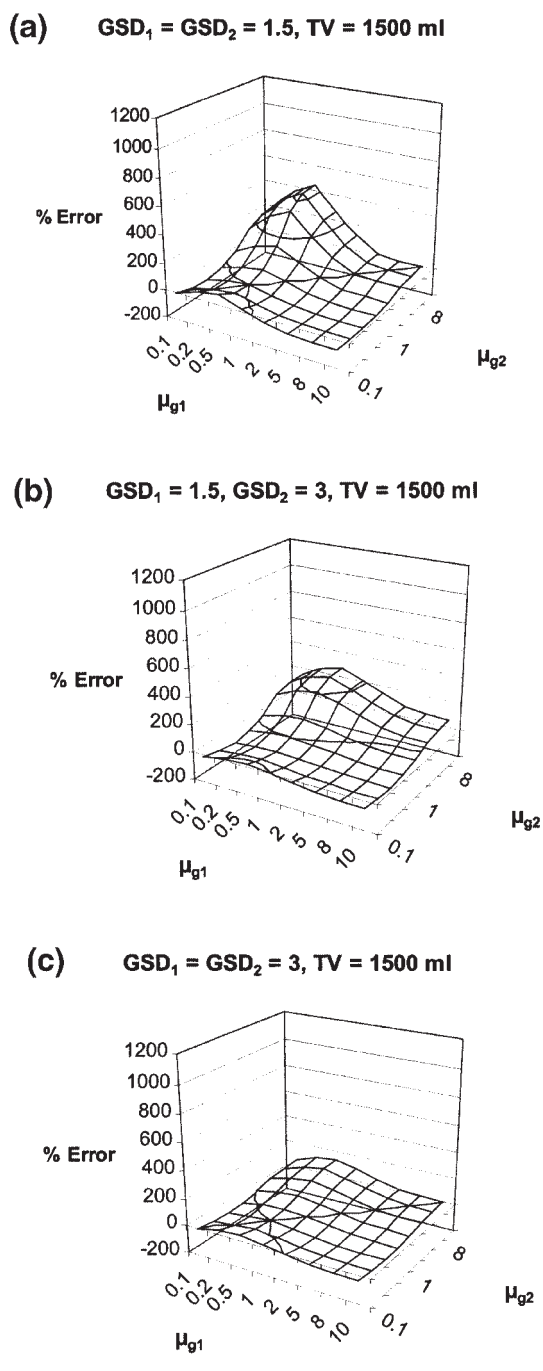

Fig. 3. Variation in percent error with μ_{g1} and μ_{g2} for breath frequency 15 min–^{1}, tidal volume 1500 cm^3 and (a) $\sigma_{g1} = \sigma_{g2} = 1.5$, (b) $\sigma_{g1} = 1.5$ and $\sigma_{g2} = 3$ or (c) $\sigma_{g1} = \sigma_{g2} = 3$.

Thus, the deposition patterns change in a manner that is similar to an increase in particle median size, consequently suggesting that the likely errors would again be closer to the maximum values for the distributions considered rather than the minimum values.

The patterns suggested by these results and the interpretation of these patterns are conclusive. This is troubling, because when a risk is estimated on the basis of aerosol penetration measurements, an *a priori* or concurrent knowledge of the aerosol particle size distribution is not necessarily assumed. With this lack of information, one can only attribute an absolute accuracy to doses that are roughly somewhere between 1/10- and 10-fold the level indicated by the

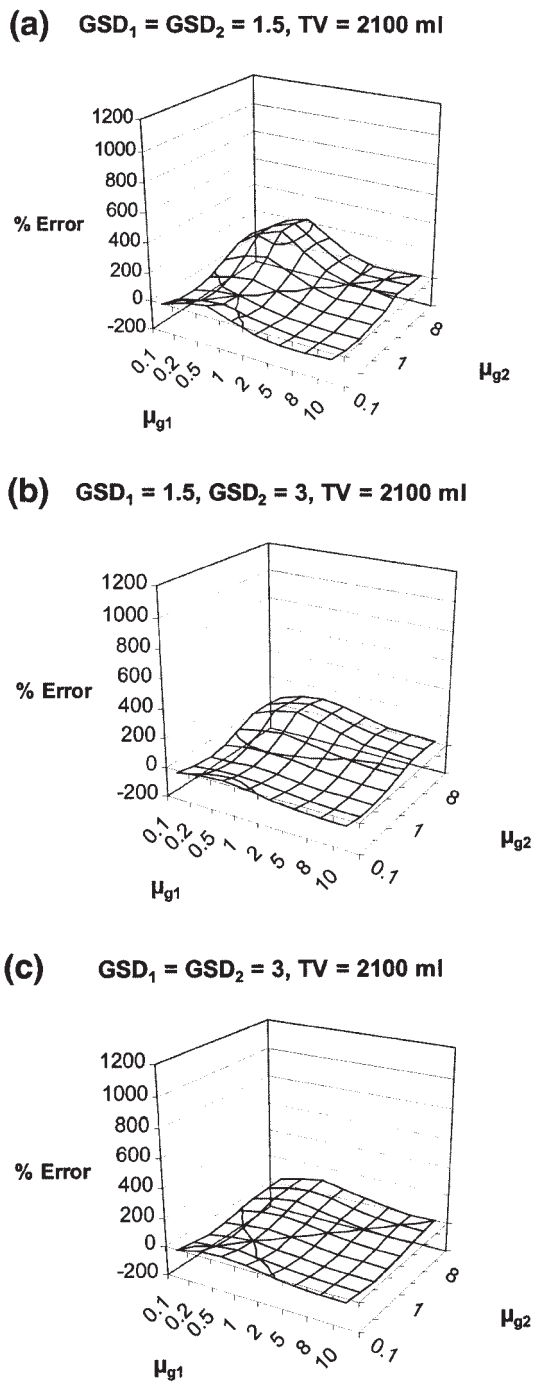

Fig. 4. Variation in percent error with μ_{g1} and μ_{g2} for breath frequency 15 min^{-1}, tidal volume 2100 cm^3 and (a) $\sigma_{g1} = \sigma_{g2} = 1.5$, (b) $\sigma_{g1} = 1.5$ and $\sigma_{g2} = 3$ or (c) $\sigma_{g1} = \sigma_{g2} = 3$.

exposure measurement, i.e. absolute dose estimation based on exposure measurement is accurate within an order of magnitude. It may be argued that the relative accuracy of the dose estimation is much better. Obviously, if the measured process does not change appreciably, then the relative merit of measurements that reflect changes in dose from day to day would constitute a consistent and accurate index for exposure-based dose. However, there would be no justification for projecting this relative merit to different processes and to some extent to the same process at different workplaces. Unfortunately, almost all of the important uses of these measurements are related to generalized populations of both workers and workplaces. Therefore, the results obtained are logically contrary to use of the measurements in determining risk for a given toxic aerosol. This observation suggests that while penetration-based measurement was an immensely important improvement over total dust measurement or unrelated dust measurement (such as impinger counts), they are not as accurate in the determination or elucidation of toxic risk as one would wish or need for the current state of hazard levels in the workplace and the environment. It is important to realize that when the undesirable consequences of exposure have very high prevalence and when the exposures are very high, even crude concordance between exposure measures and dose received are expected to yield satisfactory results. Under these circumstances, saturation of the body's defences is very likely to play an important role. However, as exposure is reduced and the undesirable health effects are also reduced to levels that are not all that much different from the background prevalence of the undesirable effect, the accuracy required for determination of exposure as a measure of dose at a critical organ increases dramatically. This would be especially true for the early detection of chronic effects. In almost all industrialized countries we are unaware of aerosol exposures that have increased dramatically over the past decades, but all of us can cite many examples of dramatic reductions in airborne particulate matter concentration for many toxic substances. Vincent and Mark (1984) pointed out the utility of using size distribution data in estimating received particulate matter doses and the ability to re-calculate received doses, at a later date, based on improved understanding of particle deposition and for differences in particle deposition with changed respiratory parameters. The current research re-emphasizes this point. In addition, the health outcome-based arguments pointed out above would suggest that if it is not currently required, the development of a deposition-based aerosol sampling methodology will soon be indispensable for the determination and quantification of inhaled aerosol-induced adverse health effects.

CONCLUSIONS

The influence of aerosol size distribution (MMAD and GSD) and breath tidal volume on respirable dose estimates were examined for mouth breathing using

Table 1. Maximum positive and negative errors calculated for 15 min^{-1} breathing rate

σ_{g1}	σ_{g2}	Maximum errors (%)					
		750 ml breath volume		1500 ml breath volume		2100 ml breath volume	
		Negative	Positive	Negative	Positive	Negative	Positive
1.5	1.5	–91	1014	–82	446	–72	261
1.5	3	–79	634	–71	272	–65	151
3	3	–68	208	–58	137	–50	101

Soderholm respirable-equivalent aerosols. Deposition estimate errors ranged from ~1/10- to 10-fold, with MMAD, GSD and tidal volume each showing a substantial influence under appropriate conditions. These findings demonstrate that reliance on respirable aerosol sampling data obtained with devices performing on a penetration-based sampling criterion may lead to erroneous dose–response relationships in exposure standard development, as well as exposure misclassification errors during epidemiological studies. A more reliable dose estimate would be obtained using devices with a collection efficiency performance closely matching the alveolar deposition curves of Heyder and Rudolf, however, given the difficulty in designing samplers that match even the relatively simple ACGIH/ISO/CEN Soderholm criterion, it seems unlikely that sampler performance based on the more complex Heyder–Rudolf relation could be achieved with simplicity. While the users of respirable air sampling data should be aware of the limitations of the data in estimating actual tissue doses, and apply appropriate caution when using these data for risk estimation and standards development purposes, it is important to start research that will lead to the development of such a methodology. Even a simple knowledge of particle size distribution would be very beneficial as research progresses to the development of a more sophisticated instrument. Reasonably detailed particle size distribution data would provide long-term flexibility in accurately estimating doses to respiratory tract tissues under various exposure conditions. We believe that if it is not currently required, the development of a deposition-based aerosol sampling methodology will soon be required for the determination and quantification of inhaled aerosol-induced adverse health effects.

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